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54 **Packaging material.**

57 A packaging material is composed of a bottom material of a first thermoplastic resin sheet having a concave portion thermally bonded therein and a cover material of a second thermoplastic resin sheet or aluminium foil which is bonded to the bottom material to seal in the said concave portion the content thereof. The bottom material consists of a polyarylene sulfide sheet or a laminate sheet having at least one layer of a polyarylene sulfide.

## Description

## PACKAGING MATERIAL

The present invention relates to a packaging material which is composed of a bottom material of a thermoplastic resin sheet or film (hereinafter, referred to as "sheet") having a concave portion formed therein and a cover material of a thermoplastic resin sheet or an aluminum foil which is bonded to the bottom material to seal in the concave portion the content thereof.

In conventional packages of tablets or the like, a PTP packaging (Press Through Pack) system in which the content comes out of the wrapping wall when the package is pressed with a finger is in general use in place of strip packaging systems.

Packages of tablets or granules are mainly used in the field of medicine. Most of the tablets are sugar-coated and most of the granules are encapsulated with gelatin so as to make the medicine easy to swallow. This is a disadvantageous, however. The sugar or gelatin can easily absorb moisture. When it absorbs moisture, not only can not the object of the packaging be achieved but also some medicines may be damaged.

Packaging such tablets of medicine or the like is considered to require a water vapor permeability of not more than  $1.0 \text{ g/m}^2 \cdot 24 \text{ hr}$  (at  $40^\circ \text{C}$  and  $90\% \text{ RH}$ ) empirically. In order to satisfy this condition, so far, sheets of aluminum, polyvinyl chloride, polyvinylidene chloride, polypropylene, polyethylene, etc. or their laminate sheet, which are materials having an excellent water vapor barrier property, are used for packaging the tablet medicine or the like [see, for example, Japanese Patent Application Laid-Open (KOKAI) Nos. 58-217,344 (1983), 60-6,452(1985) and 60-40,245 (1985)].

An aluminum foil, which has an excellent water vapor barrier property, should have thickness of not less than  $10 \mu\text{m}$ , because if its thickness is less than  $10 \mu\text{m}$ , pin holes are quite likely to exist on the foil and further, it is necessary to coat a heat sealable material on the foil in order to make the foil heat sealable with a bottom sheet. Accordingly, the foil has a deficit to be expensive.

To satisfy the water vapor barrier property, a composite sheet consisting of several laminated layers having a basic composition, such as, multiple layers of polyvinyl chloride/polyvinylidene chloride; polypropylene/polyvinylidene chloride; polyvinyl chloride/polyvinylidene chloride/polypropylene; etc. is generally used. Such composite sheet is excellent in its transparency, but since it must be used in the form of a composite sheet formed by laminating multiple layers, the manufacturing process becomes complicated and expensive.

The present inventors have extensively studied to obtain a heat-sealable packaging material which is excellent in its water vapor barrier property and transparency. Finally, it has been found that a sheet of a polyarylene sulfide or sheet of a polyarylene sulfide laminated with another thermoplastic resin sheet is a very excellent packaging material and the present invention has been achieved on the basis of this finding.

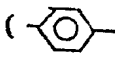


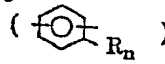
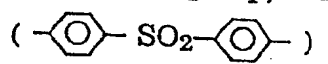
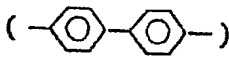
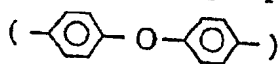
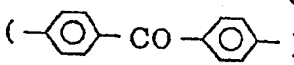
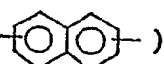
The object of the present invention is to provide a packaging material which is composed of a bottom material of a thermoplastic resin sheet with a concave portion thermally formed, and a cover material of a thermoplastic resin sheet or an aluminum foil which is bonded to the bottom material to seal a content which is put into the concave portion, the packaging material being characterized in that the bottom material consists of a polyarylene sulfide sheet or a laminate sheet having at least one layer of a polyarylene sulfide.

Another object of the present invention is to provide a packaging material in which the polyarylene sulfide is polyphenylene sulfide.

Still another object of the present invention is to provide a packaging material which makes it possible to observe its content from outside and prevents any damage to the content, particularly, due to moisture.

The present invention relates to a package which uses a polyarylene sulfide sheet or a laminate sheet having at least one layer of a polyarylene sulfide as the bottom material.

The term "polyarylene sulfide" in the present invention means a polymer having a repeating unit of  $-(\text{Ar}-\text{S})-$  (wherein Ar represents an arylene group) as the main constituent. As an arylene group, paraphenylene group

() , metaphenylene group () , orthophenylene group () ,  
 alkylsubstituted phenylene group () (wherein R represents  
 an alkyl group, preferably a lower alkyl group, and n is an integer of 1 to 4), p,p'-  
 diphenylene sulfone group () , p,p'-biphenylene group  
 () , p,p'-diphenylene ether group () ,  
 p,p'-diphenylene carbonyl group () , naphthalene  
 group () , can be used.

Among these polyarylene sulfides, polyphenylene sulfide (hereinafter, referred to as "PPS") is the most preferable in respect to physical properties such as humidity resistance, moldability and mechanical properties. In the present invention, polyparaphenylene sulfide means not only a paraphenylene sulfide homopolymer but also a random or block copolymer in which the paraphenylene group are not less than 60 mol%, preferably not less than 75 mol% of the total arylene groups. As the arylene groups other than the

As a polyarylene sulfide (hereinafter, referred to as "PAS"), a PAS, having a melt viscosity of 1,000 to 10,000 poise, preferably 2,000 to 8,000 poise measured at a temperature of 310°C and a shear rate of 200 second<sup>-1</sup>, is generally used and a non cross-linked and substantially linear polymer is preferable. Such a PAS can be produced basically by a method described in Japanese Patent Application Laid-Open (KOKAI) Nos. 61-7,332 (1986) and 61-66,720 (1986).

As the bottom material, a sheet of the PAS molded to a thickness of 30 to 1,000 μm, preferably 50 to 500 μm with an orientation as little as possible, is preferably used.

A strongly oriented sheet shrinks too much when heated for forming a bottom sheet and makes it difficult to obtain a clean one. When the sheet (strongly oriented) is heat-treated to reduce the shrinkage, its molding temperature must be higher than its heat-treating temperature and a high temperature at molding is not preferable.

The PAS sheet can optionally be used in a form of laminate sheet with a thermoplastic resin sheet as is described below. Namely, one side of the PAS sheet is oxidized, usually with corona discharge. A polyurethane adhesive or a polyester adhesive is coated on the oxidized surface to a thickness in the range of 0.5 to 5 μm by appropriately arranging its coating weight. At least one sheet of a thermoplastic resin of 10 to 500 μm in thickness, preferably 20 to 200 μm in thickness, having a surface oxidized, is laminated on the surface coated with adhesive. The thermoplastic resin is selected from the group consisting of, for example, polypropylene, polyethylene, ethylene-vinyl acetate copolymer, saponified ethylene-vinyl acetate polymer, polystyrene and polyvinyl chloride. Alternatively, a laminate sheet coated with a latex of a vinylidene chloride copolymer on the adhesive layer in a thickness of 2 to 50 μm, preferably 3 to 20 μm by solid content can be preferably used. A laminate sheet prepared by coextruding a PAS and the above thermoplastic resin can also be used.

Such a PAS sheet or PAS laminate sheet is heated to a temperature in a range of the glass transition temperature to the melting point of the PAS, namely, to a temperature of 85 to 270°C, preferably 100 to 200°C, to form a concave portion under a vacuum at a draw ratio of not more than 5. The "draw ratio" is represented by the ratio of an inside area of the concave portion (the total area of the bottom and the side walls) to the area of the sheet portion before forming which corresponds to the concave portion. After putting a tablet of medicine or the like into the concave portion, it is covered with a sheet of thermoplastic resin, while filling the concave portion with an inert gas, if necessary, and the upper and lower sheets are bonded together by heat sealing or using an adhesive and the package is sealed airtightly.

As a cover sheet, i.e., a cover material, a sheet having the same thermoplastic resin on the surface to be bonded, as the thermoplastic resin on the surface, to be bonded, of the bottom sheet, whether the sheet is single or laminate, is used. Namely, when the bottom material is a PAS, a sheet of the PAS is preferably selected as the cover sheet, and when the bottom material is a laminate sheet, a laminate sheet having the same surface material as the surface, to be heat sealed, of the bottom material is preferably selected. The thickness of the cover sheet may be thinner than that of the bottom sheet, and a cover sheet having a thickness of 10 to 200 μm is preferably used.

The PAS cover sheet may be the same as the PAS bottom sheet (non oriented or a little oriented), but it is not necessarily a non oriented sheet. In other words, a uniaxially oriented film and a biaxially oriented film are

also usable. This is because the cover sheet is not thermally formed and is only exposed to heat for a very short time when it is heat sealed.

As the cover material, it is also possible to use an aluminum foil covered with the same material as that of the surface of the bottom sheet which is in contact with the medicine etc. or, with the material which can be heat sealed with the surface of the bottom sheet which is in contact with the medicine, etc.

A material, for example, medicine, packed in the packaging of the present invention has large advantages, namely, it can be seen at least from one side and is not damaged for a long time..

Although only one layer of a PAS sheet brings a sufficiently good results according to the present invention, it goes without saying that, the PAS sheet in the form of a laminate of two or more layers, prepared by laminating one or more layer of a thermoplastic resin on it, is also available, if necessary,

A PAS may be precolored by adding a pigment, etc. to intercept ultraviolet light, and may contain a filler, etc. in the range which does not damage the object of the present invention.

#### Examples

The effectiveness of the present invention will be explained below with reference to the following examples. The tests of the physical properties in the examples were carried out by the following methods.

#### [Water vapor barrier property]

(1) The water vapor barrier property of a sheet was measured at 40°C and 90% RH in accordance with the method of JIS Z-0208, and represented by the water vapor permeability ( $\text{g}/\text{m}^2 \cdot 24 \text{ hr}$ ). The smaller the value is, the better is the water vapor barrier property.

(2) The concave portion of a sample sheet prepared by thermal vacuum forming was filled with 0.5 to 1 g of coffee granules which were sensible to humidity and change its color (light brown to dark brown) by humidity. The package containing coffee granules was sealed by heat sealing and a change in color of the granules with time was observed at 40°C and 90% RH.

#### [Transparency]

A sheet before vacuum forming was used as a sample and the haze of the sample due to white light was measured with JIS K-6714.

#### Example 1:

A non cross-linked PPS having a melt viscosity of 5,500 poise at a temperature of 310°C and a shear rate of 200  $\text{second}^{-1}$  was used as a thermoplastic resin. The PPS was extruded through a T-die into a sheet of about 200  $\mu\text{m}$  in thickness. The water vapor permeability of the sheet was as small as  $0.8 \text{ g}/\text{m}^2 \cdot 24 \text{ hour}$ . After bringing into contact with a heated plate (150°C) for 5 seconds, the sheet was formed under vacuum using a concave metal mold having a rectangular shape with rounded corners and a size of 1 cm (depth) x 1 cm (width) x 2 cm (length). About 1 g of coffee granules were put into the concave portion of the sheet. A biaxially oriented PPS sheet of 50  $\mu\text{m}$  in thickness, which had been stretched by 3 times both in the machine direction and the transverse direction at 100°C and heat treated at 200°C, was used as the cover material. The water vapor permeability of the sheet was  $1.0 \text{ g}/\text{m}^2 \cdot 24 \text{ hr}$ . The four edges of both sheets were sealed together by an impulse heat sealer. The sample was settled in a thermohygrostat of 40°C and 90% RH to observe a color change of the coffee sample with time.

#### Example 2

The same PPS as in Example 1 was extruded through a T-die into a sheet of about 100  $\mu\text{m}$  in thickness. One side of the sheet was treated with corona discharge and coated with a polyurethane adhesive (a mixture of Takelac A-310® and Takenate A-3®: manufactured by TAKEDA Chemical Industries, Ltd.) in a thickness of 0.5  $\mu\text{m}$  by solid content. A solvent of the adhesive was removed by hot air of 80°C, and the surface was further coated with a latex of vinylidene copolymer (Krehalon Latex DO-870®, manufactured by KUREHA KAGAKU KOGYO K.K.) in a thickness of 6  $\mu\text{m}$  by solid content. After the surface was dried at 80°C for 2 minutes, it was cured at 40°C for 20 hours to obtain a bottom material. The water vapor permeability of the composite sheet was  $0.85 \text{ g}/\text{m}^2 \cdot 24 \text{ hours}$ .

A biaxially oriented PPS sheet, in the same way as in Example 1, of 40  $\mu\text{m}$  in thickness was used as a cover material. One surface of the sheet was treated with corona discharge and then, coated with the same adhesive used for the bottom material above in a thickness of 0.5  $\mu\text{m}$  and further coated with Krehalon Latex DO-870 in a thickness of 6  $\mu\text{m}$  both by solid contents. The sheet was dried and then cured at 40°C for 20 hours and its water vapor permeability was  $1.0 \text{ g}/\text{m}^2 \cdot 24 \text{ hours}$ .

The base material was heated with a heated plate (120°C) and vacuum-formed to have PPS layer outside of the concave portion. Coffee granules were put into the portion and the vinylidene chloride layers of both bottom and cover were heat sealed and obtained a sample.

#### Example 3

A sample was formed in the same way as in Example 2 except that an aluminum foil of 15  $\mu\text{m}$  in thickness which is coated with an acrylic resin, was used as the cover material.

Example 4

A PPS was extruded through a T-die into a sheet having a thickness of 100  $\mu\text{m}$ . One surface of the sheet was treated with corona discharge in the same way as in Example 2 and coated with a polyurethane adhesive (a mixture of Takelac A-130 and Takenate A-3) to a thickness of about 3  $\mu\text{m}$  by solid content and was dried by hot air of 80°C for 1 minute. To the surface, a surface, treated with corona discharge, of an unoriented polypropylene sheet of 100  $\mu\text{m}$  in thickness was brought into contact and press-bonded between rolls heated to 80°C. The obtained laminate sheet was used as a bottom material. The water vapor permeability of the material was 0.9 g/m<sup>2</sup>•24 hours.

A biaxially oriented PPS sheet of 40  $\mu\text{m}$  in thickness was used as a cover material. One surface of the sheet was treated with corona discharge and coated with the same adhesive above to a thickness of 3  $\mu\text{m}$  by solid content. To the surface, a surface, treated with corona discharge, of an unoriented polypropylene sheet of 100  $\mu\text{m}$  in thickness was brought into contact and press-bonded between rolls heated to 80°C. The water vapor permeability of the cover material was 1.0 g/m<sup>2</sup>•24 hours.

The sheet for bottom was brought into contact with a heated plate (120°C) and vacuum-formed to have the polypropylene layer came into contact with a content.

Coffee granules were put into the concave portion and the polypropylene layers of both bottom and cover were heat-sealed.

Comparative Example 1

A polyvinyl chloride sheet of 300  $\mu\text{m}$  in thickness was used as the bottom material. A water vapor permeability of the sheet was 3 g/m<sup>2</sup>•24 hours. The sheet was vacuum-forming at 120°C.

The same aluminum foil as in Example 3 was used as the cover material. After charging coffee granules into this packaging material, it was heat sealed. In the atmosphere of 40°C and 90% RH, color of the content, namely coffee, started to change to black brown on the 15th day.

Comparative Example 2

One surface of a polyvinyl chloride sheet of 100  $\mu\text{m}$  in thickness was treated with corona discharge and coated with a polyurethane adhesive (a mixture of Takelac A-130 and Takenate A-3) in a thickness of about 1  $\mu\text{m}$  by solid content and was dried by hot air of 60°C. The surface was further coated with a latex of a vinylidene chloride copolymer (Krehalon Latex DO-821 S<sup>®</sup>; produced by KUREHA KAGAKU KOGYO K.K.) in a thickness of 6  $\mu\text{m}$  by solid content and was dried by hot air of 60°C. The obtained sheet was cured at 40°C for 20 hours. Two rolls of the sheet (Sheet A) were prepared. The surface, coated with vinylidene chloride, of the Sheet A (one roll) was treated with corona discharge and coated with the same adhesive above in a thickness of 2  $\mu\text{m}$  and dried by hot air of 60°C. To the surface, a surface, treated with corona discharge, of a polyethylene sheet of 30  $\mu\text{m}$  in thickness was brought into contact and press-bonded between rolls having a temperature of not higher than 40°C. The polyethylene surface of the thus-laminated sheet was treated with corona discharge and designated as (Sheet C). The surface, coated with the vinylidene chloride copolymer, of the Sheet A (the other roll), was coated with the same adhesive above in a thickness of 2  $\mu\text{m}$  by solid content and designated as (Sheet B). The polyethylene layer of the composite sheet (Sheet C) and the adhesive layer of the latter sheet (Sheet B) were brought into contact and press-bonded between rolls having temperatures of 40°C and 80°C, respectively. Consequently, a laminate sheet of five layers, namely, layers of polyvinyl chloride, the vinylidene chloride copolymer, polyethylene, the vinylidene chloride copolymer, and polyvinyl chloride laminated in that order was prepared. The water vapor permeability of the laminate sheet (5 layers) was 0.9 g/m<sup>2</sup>•24 hours and this sheet was used as the bottom material.

The obtained bottom material was heated by a heated plate (120°C), and vacuum-formed. Coffee granules were put into the concave portion and the package sample was sealed with the same aluminum foil as used in Example 3 as the cover material by heat sealing.

The results of the tests of each sheet obtained in Examples 1 to 4 and Comparative Examples 1 and 2 are shown in Table 1 together with the composition, water vapor permeability and transparency.

As is obvious from the results, the packaging material of the present invention has a very simple composition and is excellent both in transparency and in water vapor barrier property. Therefore, although PPS itself is rather expensive comparing with other resins, the packaging material of the present invention is good enough in practical use even costwise.

Table 1. Composition of Composite Sheets and Their Test Results

Ex. No.	Material for Bottom					Material for Cover			Time for color change (day)*
	Composition	Thickness (μm)	No. of Layers	Water Vapor Permeability	Transparency (%)	Composition	Thickness (μm)	No. of Layers	
Example 1	PPS	200	1	0.8	1.2	O-PPS	50	1	45
Example 2	PPS/PVDC	100/6	2	0.85	0.6	O-PPS/PVDC	40/6	2	60
Example 3	PPS/PVDC	100/6	2	0.85	0.6	Al	15	1	65
Example 4	PPS/CPP	100/100	2	0.9	4.7	O-PPS/CPP	40/100	2	50
Comp. Example 1	PVC	300	1	3.0	2.0	Al	15	1	15
Comp. Example 2	PVC/PVDC/PE/PVDC/PVC	100/6/30/6/100	5	0.9	4.0	Al	15	1	55

\* Days passed until the color of coffee changed.

PPS: Polyphenylene sulfide sheet. PVC: Polyvinyl chloride sheet

O-PPS: Biaxially oriented polyphenylene sulfide sheet. CPP: Unoriented polypropylene sheet

Al: Aluminum foil (with an acrylic resin applied). PE: Polyethylene sheet

PVDC: Vinylidene chloride copolymer.

## Claims

1. A packaging material which is composed of a bottom material of a first thermoplastic resin sheet having a concave portion thermally formed therein and a cover material of a second thermoplastic resin sheet or of an aluminium foil which is bonded to the bottom material to seal in the said concave portion the content thereof; characterised in that the bottom material consists of a polyarylene sulfide sheet or a laminate sheet comprising at least one layer of a polyarylene sulfide.
2. A packaging material according to claim 1, wherein the cover material and the bottom material have the same composition.
3. A packaging material according to claim 1 or 2, wherein the or each said polyarylene sulfide is a non cross-linked and substantially linear polymer.
4. A packaging material according to any one of the preceding claims, wherein the bottom material has a thickness of from 30 to 1,000  $\mu\text{m}$ .
5. A packaging material according to any one of the preceding claims, wherein the or each said polyarylene sulfide is a polyphenylene sulfide.
6. A packaging material according to claim 5, wherein the polyphenylene sulfide is a polyparaphenylene sulfide homopolymer or a copolymer in which not less than 75 mol% of the total arylene groups are the paraphenylene group.
7. A packaging material according to any one of the preceding claims, wherein the cover material has a thickness of from 10 to 200  $\mu\text{m}$ .
8. A packaging material according to any one of the preceding claims, wherein a tablet of a medicine is provided in the said concave portion.
9. A process for the preparation of a packaging material as claimed in any one of the preceding claims, which process comprises heating a polyarylene sulfide sheet or a laminate sheet comprising at least one layer of a polyarylene sulfide to a temperature from the glass transition temperature to the melting point of the polyarylene sulfide and forming the said concave portion therein under a vacuum at a draw ratio of not more than 5; placing the desired content of the packaging material in the said concave portion; covering the said concave portion with the said cover material, if necessary while filling the said concave portion with an inert gas; and bonding the said bottom material and the said cover material together.
10. A packaging material comprising a bottom material composed of a thermally formed polyarylene sulfide sheet or a thermally formed laminate sheet having at least one layer of a polyarylene sulfide.

(19)



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Office

# EUROPEAN SEARCH REPORT

Application Number

EP 88 30 3118

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X	EP-A-0 166 243 (UNION CARBIDE CORP.) * Page 5, lines 4-18; page 7, lines 14-20; page 54, line 1 - page 55, line 10; page 58, lines 4,5,8-18; page 63, line 29 - page 64, line 1 *	1,3-6,9 ,10	B 65 D 85/56 B 32 B 27/28
A	FR-A-2 201 671 (DAI NIPPON PRINTING CO., LTD) * Page 2, lines 25-37; page 8, line 33 - page 9, line 8; page 9, lines 23-27; figure 13 *	1,8,9	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			B 32 B
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 31-01-1989	Examiner IBARROLA TORRES O.M.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document	